2. INTRODUCTION

This document is an update revision of "Air Quality Criteria for Particulate Matter and Sulfur Oxides" published by the United States Environmental Protection Agency (EPA) in 1982, an Addendum to that document published in 1986, and an Acid Aerosols Issue Paper published in 1989, and it will serve as the basis for reevaluating the current National Ambient Air Quality Standard (NAAQS) for particulate matter (PM) set in 1987. The present document, Air Quality Criteria for Particulate Matter, critically assesses the latest scientific information relative to determining the health and welfare effects associated with exposure to various concentrations of PM in ambient air. The document is not intended as a complete and detailed literature review, but it does evaluate thoroughly information relevant to PM NAAQS criteria development based on pertinent literature available through February, 1996.

2.1 LEGISLATIVE REQUIREMENTS

Two sections (Sections 108 and 109, U.S. Code, 1991) of the U.S. Clean Air Act (CAA) govern the establishment, review, and revision of National Ambient Air Quality Standards (NAAQS). Section 108 directs the Administrator of the U.S. Environmental Protection Agency (EPA) to list pollutants that may reasonably be anticipated to endanger public health or welfare and to issue air quality criteria for them. The air quality criteria are to reflect the latest scientific information useful in indicating the kind and extent of all exposure-related effects on public health and welfare that may be expected from the presence of the pollutant in ambient air.

Section 109 directs the Administrator of EPA to propose and promulgate "primary" and "secondary" NAAQS for pollutants identified under Section 108. Section 109(b)(1) defines a primary standard as a level of air quality, the attainment and maintenance of which, in the judgment of the Administrator, based on the criteria and allowing for an adequate margin of safety, are requisite to protect the public health. Section 109(d) of the CAA requires the periodic review and, if appropriate, revision of existing criteria and standards. Under Section 109(b) of the CAA, the Administrator must set secondary NAAQS that are based on the criteria and are requisite to protect the public welfare from any known or anticipated adverse effects associated

with the presence of such pollutants. Welfare effects are impacts of air pollution not directly affecting human health, such as effects on vegetation, crops, soils, water, animals, manufactured materials, weather, visibility, and climate, as well as damage to and deterioration of property, hazards to transportation, and effects on economic value and personal comfort and well-being.

2.2 REGULATORY BACKGROUND

"Particulate matter" is the generic term for a broad class of chemically and physically diverse substances that exist as discrete particles (liquid droplets or solids) over a wide range of sizes. Particles originate from a variety of stationary and mobile sources. They may be emitted directly or formed in the atmosphere by transformation of gaseous emissions such as sulfur oxides (SO_x), nitrogen oxides (NO_x), and volatile organic substances. The chemical and physical properties of PM vary greatly with time, region, meteorology, and source category, thus complicating the assessment of health and welfare effects. Particles in ambient air usually occur in two somewhat overlapping bimodal size distributions: (1) fine (diameter less than 2.5 μm) and (2) coarse (diameter larger than 2.5 μm). The two size fractions tend to have different origins and composition.

On April 30, 1971 (Federal Register, 1971), EPA promulgated the original primary and secondary PM NAAQS under Section 109 of the CAA. The reference method for measuring attainment of these standards was the "high-volume" sampler (Code of Federal Regulations, 1986), which collects PM up to a nominal size of 25 to 45 μm (so-called "total suspended particulate" or "TSP"). Thus, TSP was the original indicator for the PM standards. The primary standards for PM (measured as TSP) were 260 $\mu g/m^3$, 24-h average not to be exceeded more than once per year, and 75 $\mu g/m^3$, annual geometric mean. The secondary standard (measured as TSP) was 150 $\mu g/m^3$, 24-h average not to be exceeded more than once per year.

On October 2, 1979 (Federal Register, 1979a), EPA announced that it was in the process of revising the Air Quality Criteria Document (AQCD) and reviewing the existing PM NAAQS for possible revisions. External review drafts of that revised AQCD were made available for public comment and peer review by the Clean Air Scientific Advisory Committee (CASAC) of EPA's Science Advisory Board (SAB). CASAC prepared a "closure" memorandum to the Administrator indicating its satisfaction with the final draft of the AQCD. After closure, minor

technical and editorial refinements were made to the AQCD (U.S. Environmental Protection Agency, 1982). The final draft of the document was issued simultaneously with the proposal of revisions to the PM standards.

On March 20, 1984 (Federal Register, 1984), EPA proposed a number of revisions to the primary and secondary PM standards. Following publication of the proposal, EPA held a public meeting in Washington, DC on April 30, 1984, to receive comments on the proposed standards revisions. After the close of the original public comment period (June 5, 1985), CASAC met on December 16 and 17, 1985, to review the proposal and to discuss the relevance of certain new scientific studies on the health effects of PM that had emerged since CASAC completed its review of the AQCD and staff paper in January 1982. Based on its preliminary review of these new studies, CASAC recommended that EPA prepare separate addenda to the AQCD and staff paper to evaluate relevant new studies and to discuss their potential implications for standard-setting. The EPA announced acceptance of these recommendations on April 1, 1986 (Federal Register, 1986a). On July 3, 1986, EPA announced (Federal Register, 1986b) the availability of an external review draft document, entitled Second Addendum to Air Quality Criteria for Particulate Matter and Sulfur Oxides (1982): Assessment of Newly Available Health Effects Information (U.S. Environmental Protection Agency, 1986). At the same time (on July 3 1986), EPA announced a supplementary comment period to provide the public an opportunity to comment on the implications of the new studies and addenda for the final standards. On October 15 and 16, 1986, the CASAC held a public meeting to review the AQCD addendum, at which time CASAC members and representatives of several organizations provided critical review comments on the subject addendum.

The CASAC sent a closure letter on the EPA AQCD addendum to the Administrator dated December 15, 1986, which stated that the 1986 addendum and the 1982 AQCD, previously reviewed by CASAC, represented a scientifically balanced and defensible summary of the extensive scientific literature on PM and SO_x (Lippmann, 1986b).

On July 1, 1987 (Federal Register, 1987), EPA published final revisions to the NAAQS for PM. The principal revisions in 1987 included (1) replacing TSP as the indicator for the ambient standards with a new indicator that includes only particles with an aerodynamic diameter less than or equal to a nominal 10 μm ("PM_{10}"), (2) replacing the 24-h primary TSP standard with a 24-h PM_{10} standard of 150 $\mu g/m^3$, (3) replacing the annual primary TSP standard with an annual

PM_{10} standard of 50 $\mu g/m^3$, and (4) replacing the secondary TSP standard with 24-h and annual PM_{10} standards identical in all respects to the primary standards.

2.3 SCIENTIFIC BASIS FOR THE EXISTING PARTICULATE MATTER STANDARDS[1]

The following discussion describes the bases for the existing PM NAAQS set in 1987. The discussion includes the rationale for the primary standards, the pollutant indicator for particulate matter, the averaging time and form of the standard, and finally a discussion of EPA's assessment that led to the standard set in 1987.

2.3.1 Rationale for the Primary Standards

In selecting primary standards for PM, the Administrator must specify (1) the particle size fraction that is to be used as an indicator of particulate pollution, (2) the appropriate averaging times and form(s) of the standards, and (3) the numerical levels of the standards. Based on the assessment of relevant scientific and technical information in the earlier 1982 PM AQCD and addenda, the staff paper and staff paper addendum outlined a number of key factors considered in making decisions in each of these areas. The following discussion of the 1987 revisions of the standards focuses mainly on the considerations that were most influential in the Administrator's selection of particular options.

[1]Adapted from Federal Register (1987).

2.3.2 Pollutant Indicator

Based on the assessment of the available scientific information, EPA concluded in 1987 that (1) a separate PM standard (as opposed to a combination standard for PM and SO_x) remained a reasonable public health policy choice, and (2) given current scientific knowledge and uncertainties, a size-specific (rather than chemical-specific) indicator should be used. In assessing the information in the AQCD, EPA reached several conclusions summarized as follows:

(1) Health risks posed by inhaled particles are influenced by both the penetration and deposition of particles in the various regions of the respiratory tract and the biological responses to these deposited materials. Smaller particles penetrate furthest in the respiratory tract. The largest particles are deposited predominantly in the extrathoracic (head) region, with somewhat smaller particles depositing in the tracheobronchial region; still smaller particles can reach the deepest portion of the lung, the pulmonary region.

(2) The risks of adverse health effects associated with deposition of typical ambient fine and coarse particles in the thoracic region (tracheobronchial and pulmonary deposition) are markedly greater than those associated with deposition in the extrathoracic region. Maximum particle penetration to the thoracic region occurs during oronasal or mouth breathing.

(3) The size-specific indicator for primary standards should represent those particles small enough to penetrate to the thoracic region. The risks of adverse health effects from extrathoracic deposition of typical ambient PM are sufficiently low that particles depositing only in that region can safely be excluded from the indicator.

Considering the above conclusions, other information on air quality composition, the need to provide protection for sensitive individuals who may breathe by mouth or oronasally and the similar convention on particles penetrating the thoracic region adopted by the International Standards Organization (1981), EPA staff recommended that the size-specific indicator include particles of diameters less than or equal to a nominal 10 μm "cut point" generally referred to as "PM_{10}". In terms of collection efficiency, this represents a 50% cut point or diameter (D_{50}), the aerodynamic particle diameter for which the efficiency of particle collection is 50%. With such a cut point, larger particles are not excluded entirely but are collected with substantially decreasing efficiency, and smaller particles are collected with increasing (up to 100%) efficiency. Ambient samplers with this cut point provide a reliable estimate of the total mass of suspended PM of aerodynamic size less than or equal to 10 μm. Such an indicator (PM_{10}) is

conservative with respect to health protection in that it includes all of the particles small enough to penetrate to the sensitive pulmonary region and includes approximately the same proportion of the coarse-mode fraction that would be expected to reach the tracheobronchial region. It places substantially greater emphasis on controlling smaller particles than does a TSP indicator, but it does not completely exclude larger particles from all control.

The assessment of then-available information on respiratory tract deposition in the 1986 AQCD and staff paper addenda reinforced the conclusions reached in the original EPA assessment. In particular, (1) the data did not provide support for an indicator that excluded all particles larger than 10 μm in diameter; (2) the analysis used to support an alternative indicator with a nominal size cut point of 6 μm (Swift and Proctor, 1982) significantly underestimated thoracic deposition of particles larger than 6 μm in diameter under natural breathing conditions; (3) the PM_{10} indicator generally included a similar or larger fraction of the range of particles that can deposit in the tracheobronchial region, although it appeared to be somewhat less conservative in this regard than previously thought with respect to large (>10 μm) particle deposition under conditions of natural mouthbreathing; and (4) the studies of tracheobronchial deposition generally involved adult subjects (other information indicating even greater tracheobronchial deposition of particles in children than in adults provided an additional reason for an indicator that includes particles capable of penetration to the tracheobronchial region). Consideration of these and the earlier conclusions led EPA to reaffirm its recommendation for a PM_{10} indicator. The CASAC also restated its support for PM_{10} in its review of the proposal and the closure letter to the Administrator (Lippmann, 1986a,c).

In 1987 the Administrator accepted the recommendations of the staff and CASAC, as well as their underlying rationale, and decided to replace TSP as the particle indicator for the primary standards with a new indicator that included only those particles less than a nominal 10 μm in diameter (PM_{10}) as specified in the Federal Reference Method.

2.3.3 Averaging Time and Form of the Standards

The EPA's assessment at that time of scientific information available prior to 1987 confirmed the need for both short- and long-term primary standards for PM. The alternative of a single averaging time would not provide adequate protection against potential effects from both long- and short-term exposures without being unduly restrictive. The forms for the 24-h and annual standards are discussed below.

2.3.3.1 24-Hour Standard

The Environmental Protection Agency decided in 1987 that the 24-h standard should be stated in a statistical form that uses more than 1 year of data and accounts for variations in sampling frequency in order to predict the actual number of exceedances to be expected in an average year. When used with an appropriate standard level, the statistical form can provide improved health protection that is less sensitive to changes in sampling frequency than the deterministic form and can also offer a more stable target for control programs. Recognition of the limitations of the deterministic form also led EPA to promulgate a statistical form for the ozone standard (Federal Register, 1979b).

2.3.3.2 Annual Standard

The EPA Administrator decided to change the form of the annual standard in 1987 from the previous annual geometric mean form to a statistical form expressed as an expected annual arithmetic mean. The expected annual arithmetic mean is equivalent to the long-term arithmetic average concentration level, assuming no changes in underlying emissions. The expected arithmetic mean is more directly related to the available health effects information than is the annual geometric mean, which was the previous form of the standard. Because the arithmetic mean concentration is proportional to the sum of the daily means, it reflects the total cumulative exposure of PM to which an individual is exposed. Thus, it is an appropriate indicator to protect against any health effect that depends on chronic, cumulative PM exposure. It is also a reasonable indicator for protecting against health effects that depend on repeated short-term high concentrations (short-term peaks have an influence on the arithmetic mean that is proportional to their frequency, magnitude, and duration). The geometric mean, on the other hand, deemphasizes the effects of short-term peak concentrations and is heavily influenced by days of

relatively clean air. For these reasons, EPA staff and CASAC recommended the change to an arithmetic mean.

Under the statistical form, the expected annual arithmetic average is determined by averaging the annual arithmetic averages from 3 successive years of data. The prior deterministic form of the standard did not adequately take into account the random nature of meteorological variations. In general, annual mean PM concentrations vary from year to year, even if emissions remain constant, due to the random nature of meteorological conditions that affect the formation and dispersion of particles in the atmosphere. If only 1 year of data is considered, compliance with the standard and, consequently, emission control requirements, may be determined on the basis of a year with unusually adverse or unusually favorable weather conditions. The problem of year-to-year variability is, however, reduced by averaging 3 years of data.

2.3.4 Level of the Standards

The original Office of Air Quality Planning and Standards (OAQPS) PM Staff Paper and CASAC recommendations set forth a framework for determining the levels for the proposed PM standards that would protect public health with an adequate margin of safety. The Administrator's decision in 1987 relied heavily on that framework and on the supporting material in the staff paper and its addendum, as well as the CASAC closure letters. The essential steps in this framework are summarized here.

2.3.4.1 Assessment of the Quantitative Epidemiological Studies

The 1982 AQCD and its 1986 addendum identified a small number of community epidemiological studies that are useful in determining concentrations at which PM is likely to adversely impact public health. The EPA staff used these quantitative studies to examine concentration-response relationships and to develop numerical "ranges of interest" for possible PM_{10} standards.

A number of uncertainties associated with the use of these studies had to be considered in selecting an appropriate margin of safety. As discussed in the staff paper, the AQCD, and the addenda to those documents, epidemiological studies are generally limited in sensitivity and are subject to inherent difficulties involving control for covariates or confounders. Moreover, many

of the quantitative studies were conducted in times and places where pollutant composition may have varied considerably from current U.S. atmospheres. Also, most of the studies used British Smoke—British Smoke (BS) is a pseudo-mass indicator related to small particle (aerodynamic diameter less than a nominal 4.5 μm) darkness—or TSP as particle indicators. None of the published studies used the proposed PM_{10} indicator. Thus, assumptions had to be used to convert the various results to common (PM_{10}) units.

2.3.4.2 Identification of Margin of Safety Considerations

The 1982 AQCD and its addendum identified an additional substantial body of scientific literature that, although it did not provide reliable concentration-response relationships for ambient exposures, did provide important qualitative insights into the health risks associated with human exposure to particles. This literature included both quantitative and qualitative epidemiological studies, controlled human exposure experiments, and animal toxicological studies. The EPA staff assessed this literature to identify additional factors and uncertainties that should be considered in selecting the most appropriate margin of safety.

Experience had shown that it was difficult to identify, with confidence, the lowest pollution level at which an adverse effect would occur. Furthermore, in cases such as the present one, the evidence suggested that there is a continuum of effects, with the risk, incidence, or severity of harm decreasing, but not necessarily vanishing, as the level of pollution is decreased.

The requirement for an adequate margin of safety for primary standards addresses uncertainties associated with inconclusive scientific and technical information available at the time of standard setting. It also aims to provide a reasonable degree of protection against hazards that research has not yet identified. Both kinds of uncertainties are components of the risk associated with pollution at levels below those at which human health effects can be said to occur with reasonable scientific certainty. Thus, by selecting primary standards that provide an adequate margin of safety, the Administrator sought not only to prevent pollution levels that have been demonstrated to be harmful, but also to prevent lower pollutant levels that may pose an unacceptable risk of harm, even if that risk is not precisely identified as to nature or degree.

In the absence of clearly identified thresholds for health effects, the selection of a standard that provides an adequate margin of safety requires an exercise of informed judgment by the Administrator. The level selected will depend on the expected incidence and severity of the

potential effects and on the size of the population at risk, as well as on the degree of scientific certainty that the effects will in fact occur at any given level of pollution.

The 1986 EPA staff paper recommended a range of potential standards for the Administrator's consideration. The recommended range was below the levels at which EPA staff, with the concurrence of CASAC, had concluded from the available data that adverse health effects were "likely", but in the domain where the data suggested that such effects were "possible". The Administrator proposed refined ranges of standard levels that were based on the 1984 staff and CASAC recommendations. After consideration of the then new scientific evidence contained in the AQCD addendum, the staff revised its recommended range of standards. The Administrator considered the revised EPA assessments and the CASAC recommendations (Lippmann, l986b) in making the final decision on the standard levels. The rationales for the levels of the 24-h and annual standards are presented below.

2.3.4.3 24-Hour Standard

The 1987 assessment of the short-term epidemiological data expressed PM levels in both the BS or TSP and PM_{10} units. The term "effects likely" denoted concentration ranges derived from the 1982 AQCD and its addendum at or above which a consensus judgment suggested the greatest certainty that the effects studied would occur, at least under the conditions that occurred in the original studies. In the "effects possible" range, EPA found credible scientific evidence suggesting the existence of adverse health effects in sensitive populations, but substantial uncertainty existed regarding the conclusions to be drawn from such evidence.

The 1987 review of the data did not provide evidence of clear thresholds in exposed populations. Instead, they suggested a continuum of response for a given number of exposed individuals, with both the likelihood (risk) of any effects occurring and the extent (incidence and severity) of any potential effect decreasing with concentration (this was particularly true for the statistical analyses of daily mortality in London). Substantial agreement existed that wintertime pollution episodes produced premature mortality in elderly and ill populations, but the range and nature of observed associations provided no clear bases for determining lowest effects-likely levels or for defining a concentration below which no association remained. The lung function studies in children also provided evidence of effects at concentrations over a range, but the relationships were not certain enough to derive effects-likely levels for PM_{10}. The lung function

studies did, however, suggest levels below which detectable functional changes were unlikely to occur in exposed populations. Following CASAC recommendations, EPA used the combined range of effects-possible studies as a starting point for developing alternative standards.

The original range proposed by the Administrator, drawn from the 1984 staff analysis, was 150 to 250 $\mu g/m^3$ PM_{10} 24-h average, with no more than one expected exceedance per year. The lower bound of this range was derived from the original assessment of the London mortality studies. As a result of reanalyses of the London mortality data and the findings from the then current U.S. morbidity studies, the staff reduced the level of the lower bound of the range of interest to 140 $\mu g/m^3$, and noted that the difference between it and the original lower bound (150 $\mu g/m^3$) was within the range of uncertainty associated with converting the morbidity study results from TSP to PM_{10}.

At that time, the study of Lawther et al. (1970) was judged to provide evidence that health effects were likely at PM concentrations above 250 $\mu g/m^3$ (as BS). The effects observed in this study (related to aggravation of bronchitis) were of concern because of both their immediate impact and their potential for inducing longer term deterioration of health status in a significant sensitive group. Based on the uncertain conversion between smoke and PM_{10}, the lowest effects likely level derived from the Lawther study (250 $\mu g/m^3$ as BS) was judged to be in the range of 250 to 350 $\mu g/m^3$ in PM_{10} units.

The 1987 assessment of the Lawther et al. (1970) study formed the basis for the upper bound of the range of PM_{10} standards proposed by the Administrator in 1984. Considering this study alone, a PM_{10} standard of 250 $\mu g/m^3$ might have appeared to contain some margin of safety, even for the sensitive bronchitics studied, because it incorporated a conservative PM_{10} conversion factor and because of differences between exposure conditions in the British study and current U.S. air quality. Because persons with chronic bronchitis were identified as a group particularly sensitive to particulate pollution, a standard of 250 $\mu g/m^3$ (as PM_{10}) also might have provided some margin of safety for other, less sensitive groups. Nevertheless, this study of bronchitics in London had inherent limitations in sensitivity that precluded derivation of unequivocal "effects thresholds" at 250 $\mu g/m^3$ as BS and, by extension, PM_{10}. The 1982 AQCD noted that associations between pollution and health status persisted at lower BS concentrations in selected, more sensitive individuals. Although the lead author of the study objected to attaching any importance to these latter findings (Lawther, 1986), EPA, with CASAC

concurrence, found no basis for asserting that this study demonstrated a population threshold at 250 $\mu g/m^3$.

In evaluating the margin of safety for a 24-h standard, it was also important to consider the London mortality studies. A standard at the upper portion of the proposed range (250 $\mu g/m^3$) would be well below the levels (500 to 1,000 $\mu g/m^3$ as BS) of the historical London episodes in which the scientific consensus indicated that pollution was responsible for excess mortality. The portions of the population at greatest risk of premature mortality associated with PM exposures in those episodes included the elderly and persons with preexisting respiratory or cardiac disease. Although the extent of life shortening could not be specified, the seriousness of the effect strongly justified a margin of safety for it (below the consensus effects levels) that was larger than that warranted for the effects on bronchitis.

The staff assessment at that time of several reanalyses of London mortality suggested, however, that the risk of premature mortality for sensitive individuals extended to concentrations substantially lower than those that occurred in the "episodes". Other analyses (Mazumdar et al., 1982; Ostro, 1984; Shumway et al., 1983) provided no objective support for a population threshold below which such a risk no longer existed. Although the risk to individuals may be small at concentrations of 250 $\mu g/m^3$ and below, the number of people exposed to lower concentrations, given U.S. levels, was substantially larger than the number exposed to higher levels. The increased number of individuals exposed increased the risk that effects would occur in the total population exposed.

Differences in the composition of particles and gases among U.S. cities and between U.S. conditions and London at the time that the mortality and morbidity data were gathered added to the complexity of assessing risk associated with PM in the United States. In the case of the mortality studies, however, the staff found that at least one study (Özkaynak and Spengler, 1985) provided qualitative support for an association between daily mortality and particle concentrations in then nearly contemporary U.S. atmospheres.

The 1982 assessment of the mortality studies and related factors prompted the EPA Administrator to consider standard levels that extended from 250 $\mu g/m^3$ to the lower bound of the original staff range (150 $\mu g/m^3$) and even lower. Reanalyses of the London mortality data prior to 1987 provided additional evidence that serious adverse health effects may occur at PM concentrations below 250 $\mu g/m^3$. These analyses addressed a number of the uncertainties

associated with the earlier studies and reinforced the Administrator's concern that a 24-h standard at the upper end of the proposed range may not provide an adequate margin of safety. However, given the uncertainties in converting from BS to PM_{10} measurements, particularly at lower concentrations and the possible differences in particle composition between London at the time the data were gathered and the contemporary United States, it was difficult to use these studies to set a precise level for a PM_{10} standard.

Given these difficulties, it was important to examine studies contemporary with the other studies that utilize gravimetric measurements of particulate concentrations. The staff found the studies of Dockery et al. (1982) and Dassen et al. (1986) to be useful. The Dockery study observed physiologically small but statistically significant decreases in lung function in a group of children exposed to peak PM_{10} levels of 140 to 250 $\mu g/m^3$. The decrements persisted for 2 to 3 weeks following the exposures. The study also suggested the possibility of larger responses in a subset of the children, including those with existing respiratory symptoms. The Dassen study recorded similar decrements in children in the Netherlands following exposure to PM_{10} levels estimated at 200 to 250 $\mu g/m^3$, but no observable effects 2 days after exposure to PM_{10} levels estimated at 125 $\mu g/m^3$. The particle composition, at least in the Dockery study, was more representative of contemporary U.S. cities in that time period, and the associated aerometry provided a more reliable estimate of PM_{10} levels than did the measurements used in the London studies. It was reasonable to expect the endpoints observed (small reversible reductions in lung function in children) to be, in most cases, more sensitive to air pollution than those observed in the London studies. These effects, *per se*, are of uncertain significance to health, but they may be associated with aggravation of respiratory symptoms in children with preexisting illness. Long-term examination of respiratory health in the same community studied by Dockery et al. (1982) suggested that the children in that community had a higher incidence of respiratory illness and symptoms than children in communities with lower particle levels, but the data showed no evidence for any persistent reduction in lung function (Ware et al., 1986). Uncertainties with respect to the effects of other pollutants (e.g., sulfur dioxide), the consistency of the changes, and exposures precluded specifying unequivocal "effects likely" levels based on this study. The EPA assessment therefore suggested that short-term lung function effects in children were possible across a range of 140 to 250 $\mu g/m^3$ or more as PM_{10}.

In making a decision on a final standard level, the Administrator also considered information from the more qualitative studies of PM. These studies suggested increased risks for sensitive groups (asthmatics) and risks of potential effects (morbidity in adults) not demonstrated in the more quantitative epidemiological literature. The qualitative studies did not provide clear information on effect levels but did justify consideration of effects of PM that have not been sufficiently investigated.

Based on the 1982 assessment of the available scientific data, in 1984, the EPA Administrator expressed an inclination to select a 24-h level from the lower portion of the proposed range of 150 to 250 $\mu g/m^3$. The addendum to the 1982 assessment supported the original findings and, if anything, suggested an even wider margin of safety was warranted. The Administrator, therefore, decided to set the final standard at the lower bound of the range originally proposed (i.e., 150 $\mu g/m^3$). This standard provided a substantial margin of safety below the levels at which there was a scientific consensus that PM caused premature mortality and aggravation of bronchitis. Such a margin was judged to be necessary because of the seriousness of the effects and because the analyses of daily mortality studies suggested that adverse effects may occur at PM levels well below the consensus levels. The standard was in the lower portion of the range where sensitive, reversible physiological responses of uncertain health significance had been possibly, but not definitely, observed in children. Using a conservative assessment of the lung function/particle relationship from Dockery et al. (1982), a change in concentration from background levels (≈ 20 $\mu g/m^3$) to 150 $\mu g/m^3$ would produce lung function changes of at most 10 to 15% in less than 5% of exposed children. Based on the results of Dassen et al. (1986), it appeared unlikely that any functional changes would be detected 1 or 2 days following such exposures. Thus, the maximum likely changes in lung function appeared to present little risk of significant adverse responses. Standards set at a somewhat higher level, however, would have presented an unacceptable risk of premature mortality and would have allowed the possibility of more significant functional changes. Furthermore, a standard level of 150 $\mu g/m^3$ was fully consistent with the recommendations of CASAC on the 24-h standard (Lippmann, 1986c).

2.3.4.4 Annual Standard

The long-term epidemiological studies examined in 1987 were subject to confounding variables that reduced the studies' sensitivity and made their interpretation difficult. No clear thresholds could be identified for the effects-likely levels, and evidence existed for effects at lower levels (the effects-possible levels); however, the evidence was inconclusive, and the effects were difficult to detect.

Based on an EPA assessment of PM_{10}/TSP ratios in areas with elevated TSP levels, the effects-likely levels from the Ferris et al. (1973) study were revised to 80 to 90 $\mu g/m^3$ as PM_{10}. Because of limitations in sampling duration and the conversion to PM_{10}, this estimate was particularly uncertain, with effects possible at lower concentrations. Of greatest concern was the possibility of long-term deterioration of the respiratory system in exposed populations, the potential for which was indicated by lung function (mechanical pulmonary) changes and increased incidence of respiratory disease. One set of studies (Ferris et al., 1973, 1976) provided some evidence for a "no-observed-effect level" for those effects at or below 60 to 65 $\mu g/m^3$ as PM_{10} (130 $\mu g/m^3$ as TSP), whereas another study (Bouhuys et al., 1978) suggested some possibility of symptomatic responses in adults at long-term median levels at or below about 50 to 55 $\mu g/m^3$ as PM_{10}. The importance of these symptomatic responses, which were unaccompanied by lung function changes, to long-term respiratory health was unclear.

The most important study of long-term effects at that time was an ongoing examination of six U.S. cities (Ware et al., 1986). The study indicated the possibility of increased respiratory symptoms and illnesses in children at multiyear levels across a range of 40 to more than 58 $\mu g/m^3$ as PM_{10} but found no evidence of reduced lung function at these concentrations. This study did not find similar gradients in symptoms and illness within some of the cities, which had somewhat smaller localized pollution gradients. The results of a separate series of studies of long- and intermediate-term (2- to 6-week) exposures in a number of U.S. metropolitan areas (Ostro, 1987; Hausman et al., 1984) were more supportive of the possibility of effects within cities (respiratory-related activity restrictions in adults) at comparable U.S. exposure levels. The results of these studies were generally consistent with the earlier U.S. studies. In particular, the finding of symptomatic responses in children with no change in lung function (Ware et al., 1986) was consistent with similar findings in adults (Bouhuys et al., 1978) at estimated long-term PM_{10} levels down to 50 $\mu g/m^3$. However, the information available to support the

existence of significant adverse effects at annual PM_{10} levels below 50 $\mu g/m^3$ (especially when 24-h levels are maintained below 150 $\mu g/m^3$) was quite limited and uncertain.

Because of the uncertainties and the limited scope and number of long-term quantitative studies available for review in 1987, it was important to examine the results of qualitative data from a number of epidemiological, animal, and ambient particle composition studies in determining what would constitute an adequate margin of safety for an annual standard. These studies justified concern for serious effects not directly evaluated in the above studies. Such effects included damage to lung tissues contributing to chronic respiratory disease, cancer, and premature mortality. Substantial segments of the population may be susceptible to one or more of these effects. Although the qualitative data did not provide evidence for major risks of these effects at the annual PM levels in most U.S. cities at that time, the Administrator believed that the seriousness of the potential effects and the large population at risk warranted caution in setting the standard.

Based on findings discussed in the 1982 AQCD, the EPA Administrator proposed in 1984 to select an annual standard level from a range of 50 to 65 $\mu g/m^3$. In the proposal, the Administrator favored a standard in the lower portion of the range. The evidence discussed in the 1986 addendum, although subject to substantial uncertainty, reinforced this inclination. In light of the 1986 assessment, and in accordance with CASAC recommendations, the Administrator decided to set the level of the annual standard at the lower bound of the original range (50 $\mu g/m^3$, expected annual arithmetic mean). This standard provided a reasonable margin of safety against long-term degradation in lung function, which was judged likely to occur at estimated PM_{10} levels above 80 to 90 $\mu g/m^3$ and for which there was some evidence at PM_{10} levels above 60 to 65 $\mu g/m^3$. Such a standard also provided reasonable protection against the less serious symptomatic effects (bronchitis) for which only inconclusive evidence was available. Moreover, the staff and CASAC recommended that the combined protection afforded by both 24-h and annual NAAQS be considered in selecting the final standard level. In this regard, analyses of air quality data showed that implementation of the 24-h standard would reduce substantially the annual levels in many U.S. areas to below 50 $\mu g/m^3$, adding to the protection afforded by the annual standard in areas with higher 24-h peak-to-mean ratios. Based on the then available information on risks associated with annual exposures, the EPA

Administrator believed that the annual and 24-h standards provided an adequate margin of safety.

2.3.5 Welfare Effects

No convincing evidence existed indicating significant adverse soiling and nuisance at TSP levels below 90 to 100 $\mu g/m^3$, and, on that basis, the Administrator concluded that secondary standards different from the primary standards were not requisite to protect the public welfare against soiling and nuisance. This conclusion was supported by CASAC's determination that there was no scientific support for a TSP-based secondary standard (Lippmann, 1986c). Therefore, the Administrator decided to set 24-h and annual secondary PM_{10} standards that are equal in all respects to the primary standards.

The other welfare effects of principal interest were impairment of visibility, potential modification of climate, and contribution to acidic deposition. All three of these effects were believed to be related to regional-scale levels of fine particles, and control programs designed to ameliorate them would likely involve region-wide reductions in emissions of sulfur oxides. The Administrator also concurred with the staff suggestions that a separate secondary particle standard was not needed to protect vegetation or to prevent adverse effects on personal comfort and well-being.

2.4 TOPICS/ISSUES OF CONCERN FOR CURRENT CRITERIA DEVELOPMENT

Based on the available scientific evidence, several critical topics and associated issues are addressed in this document, as part of the current CAA-mandated periodic review of criteria and NAAQS for PM. Some of the most critical topics and issues addressed are as follows.

2.4.1 Air Quality and Exposure

2.4.1.1 Physics and Chemistry of Atmospheric Aerosols

The atmospheric aerosols of interest because of their potential health and welfare effects consist of two principal components: a gas phase ("air" in this case) and a solid or liquid particle phase in suspension. Fine particles in the atmosphere consist mainly of (1) sulfate, nitrate,

ammonium ions, and water; (2) photochemically formed organic aerosols; and (3) carbon, organic matter, and metallic components emitted directly into the atmosphere. Coarse particles in the atmosphere are composed mainly of silica, calcium carbonate, clay minerals, soot, and, sometimes, organic substances. A general relationship exists between chemical composition and particle diameter, with particles of ≤ 2.5 μm in diameter containing most of the SO_4^{2-}, H^+, and NH_4^+, as well as a significant fraction of the NO_3^- and Cl^-. The particle volume (mass) frequency function is often multimodal. The fine-volume fraction may have two or more modes below 1.0. The coarse fraction generally has one mode within the range ≈ 5 to 50 μm. The particle volume frequency functions for the fine and coarse fractions often behave independently, (i.e., vary in relative proportion of the total ambient particle mix from location to location or from one time or season to another at the same location).

Previous documentation has shown that hydroxy, hydroperoxy, and alkoxy radicals are probably important in the oxidation of SO_2 to SO_3^-, although the rate constants for some of these reactions are not well established. The hydroxy radical dominates the gas-phase oxidation of SO_2 in the clean troposphere, and H_2O_2 is effective in the formation of SO_4^{2-} in particles, mists, fogs, and rain. Transition metals and soot have been shown to be effective catalysts for atmospheric oxidation of SO_2. Oxidation rates for NO and NO_3^- are known but have been considered too low to be important. The oxidation rate for NO_2^- is known, but the tropospheric concentration of HNO_2 is probably too low for this reaction to be important. Except for reactions of carbon (soot), solid surface reactions do not appear to be effective pathways for H_2SO_4 formation in the troposphere.

The physical properties of particles are physical configuration, bulk material properties, and surface properties. The bulk material properties that affect aerosol behavior include chemical composition, vapor pressure, hygroscopicity and deliquescence, and index of refraction. These properties control the physical state and growth of particles and result in scattering and absorption of light by tropospheric particles. Hygroscopicity, deliquescence, and efflorescence are critical properties in the growth of particles, but there is a paucity of thermodynamic data to permit prediction of deliquescence and hygroscopic behavior and vapor pressures of multicomponent systems, especially for relative humidities below about 90%. Few studies of desorption under atmospheric conditions have been reported; of more concern, desorption may prove to be important in biological systems. Shape, structure, and density are

physical configuration properties that are important parameters in the equations of motion for particles. Because of irregularities in particle geometry or because the particles are agglomerates, the three configuration properties are usually defined in terms of an aerodynamic diameter. Surface properties of importance include electrostatic charging, adhesion, and the influence of surface films.

The physical properties of particles and their modal distributions are important considerations (1) in the sampling and analysis of atmospheric particles and (2) in predicting or determining the flux to biological and nonbiological materials and deposition in the human and laboratory animal respiratory tracts.

Advances in understanding the properties and behavior of atmospheric particulate matter have been made since publication of the previous criteria document (U.S. Environmental Protection Agency, 1982). In the current revision of the document, newer literature and data on the above topics are reviewed and discussed. For example, chemical pathways and rates of atmospheric particle formation and of removal from the atmosphere, by dry deposition and by precipitation scavenging, are examined. Likewise, the physical processes of nucleation, condensation, and coagulation by which condensible material is converted into particles are discussed, along with the size distribution of the resulting particles. The physical properties relevant to sampling considerations and deposition on surfaces, including those of the respiratory tract, are also discussed, including coverage of several newer areas of expanded research: aerosol equilibria, the unique properties of semi-volatile aerosols, and the role of particle-bound water.

2.4.1.2 Measurement Methodology

Techniques available for measurement of mass and specific components of aerosols are examined. Special attention is given to the suitability of current technology for measurement of aerosol mass with sufficient accuracy and precision to determine compliance with one or another possible type of a new PM standard (i.e., a PM_{10} standard with a lower level or a fine-particle standard). The need for continuous or daily PM measurements, the difficulty of removing particle-bound water without losing NH_4NO_3 or semivolatile organic matter, and problems in defining and maintaining a precise cut at 10 μm or lower (e.g., at 2.5 μm) are also assessed.

2.4.1.3 Ambient Levels

The present draft of the revised PM AQCD describes ambient PM data for the United States, with characterization as available by size (fine/coarse) and chemical composition. Data that focus on the current U.S. PM_{10} standard are emphasized, but information is also provided on $PM_{2.5}$, $PM_{2.5-10}$, and other similar cut points, as data are available. Ambient patterns are discussed, to include daily, seasonal, regional, etc. Acid aerosol data are also described as above as a separate aspect of PM. Key questions addressed include: What information is available on the distribution of PM with regard to: geographic, seasonal, diurnal, size, composition, sources, and trends? How important are uncertainties introduced by variations in the position and shape of the 10-μm cut point in various PM_{10} monitors? How important are measurement uncertainties due to volatilizable/condensible components (e.g., loss of ammonium nitrate and, possibly, other ammonium salts) or to the loss of semivolatile organics or retention of particle-bound water? How do these uncertainties vary geographically and seasonally? How do these uncertainties differ for filter collection and subsequent weighing as compared to continuous indicators?

2.4.1.4 Cut Points

Information helpful in evaluating the possible need for a new fine particle standard in addition to or instead of a PM_{10} NAAQS is presented. This information includes discussion of sources, sampling problems, composition, lung deposition, epidemiology, biochemistry, and toxicology of fine and coarse particles. Other considerations include techniques for separating fine particles from coarse particles. Can fine and coarse particles be separated adequately by a single size cut-point in all areas of the country or will the optimal cut point differ in time and space, especially between very dry areas where coarse particles may be found below 2.5 μm and very humid areas where fine particles occur above 1.0 μm? If a single fine-particle cut point is chosen, which is best: 2.5 μm; 1.0 μm; or something in between? Is separation by size adequate or will chemical composition measurements also be needed?

2.4.1.5 Exposure

Particulate matter exposure estimates for most epidemiology studies are based on data from ambient monitoring sites. Relationships between such measurements and personal exposure are of interest in evaluating and interpreting epidemiology studies. Aspects assessed in

the present document include: urban scale PM exposure models, indoor/outdoor PM characteristics and relationships, and the validity of ambient measurements to provide appropriate estimates to relate to health effect endpoints. Two exposure estimates are of concern, individual and population estimates of PM exposure. The type of epidemiology study determines which estimate is appropriate. Additionally, other factors (such as exposure durations) that may determine health effects are considered. Human exposure patterns to ambient and indoor air particles, including consideration of activity patterns and various microenvironments, are also characterized.

Actual human exposure differs from outdoor concentrations due to: the infiltration of ambient aerosols indoors; indoor sources; and human activity patterns. Human exposure can be determined through measurements and models. For PM, indoor and personal monitoring data show both higher than ambient and lower than ambient PM concentrations in indoor settings as a function of varying particle size and human activity patterns.

Coarse-mode particles (>2.5 μm), which are generally of nonanthropogenic origin (windblown dust, etc.), require turbulence to provide vertical velocity components greater than their settling velocity to allow them to remain suspended in the air. Outdoor particles enter into an indoor setting either (1) by bulk flow (e.g., through an open window) in which all particles can enter at the inlet condition or, (2) by diffusional flow (e.g., through cracks and fissures in the barrier of the building envelope). Current investigations suggest that both fine and coarse particles penetrate indoors with high efficiency. However, indoor settings are usually quiescent, and the larger ambient particles that do enter indoors quickly settle out, leading to the presence of the familiar dust layers that require indoor settings to be cleaned constantly. Fine particles, which enter indoors, however, are not easily removed by settling or impaction and are more reflective of ambient fine particle concentrations than are coarse particles. Human activity in indoor settings does generate fine particles (<2.5 μm) from smoking, vacuuming, cooking, etc., and resuspends coarse particles that previously had settled out. Thus, indoor PM consists of both: (a) ambient particles which have penetrated indoors and remain suspended, and (b) particles generated indoors.

Two major factors influencing the relationship of ambient to indoor PM air quality are (1) the variability of indoor concentrations of PM compared to outdoor concentrations as a function of particle size (e.g., fine indoor $\geq$ fine outdoor and coarse indoor < coarse outdoor) and (2) the

variation of exposures of individuals related to the different activities that are involved with the local generation of particles in their immediate surroundings (smoking, traffic, dusting and vacuuming at home, etc.).

Long-term personal exposures to coarse-fraction PM (>2.5 μm) can be less than half the ambient concentrations. Long-term personal exposures to fine-fraction PM (<2.5 μm) of ambient origin may be estimated by ambient measurements of the <2.5 μm PM fraction. However, the concentration of particles generated indoors or due to personal activities would not be expected to vary in concert with ambient concentrations. Therefore, variations in ambient concentrations can serve as an indicator of variations in total exposure to ambient particles, experienced both outdoors and in various microenvironments.

2.4.2 Health Effects

A rapidly growing body of epidemiologic data examines associations between PM concentrations and human health effects, ranging from respiratory function changes and symptoms to exacerbation of respiratory disease and excess mortality. These effects appear to lie along an increasing gradient of severity of effects in different subpopulations. Although the exact biological mechanisms underlying such effects are poorly understood, the emerging pattern of findings points toward the plausibility that the observed associations likely reflect real relationships between ambient PM exposures and human health impacts. This revised PM criteria document assesses evidence suggesting that this overall pattern of effects may extend to concentrations of PM_{10} below the current NAAQS or may be associated with other PM size fractions (e.g., fine particles < 2.5 μm). Controlled human exposure and laboratory animal studies are also evaluated, and the overall coherence and consistency of findings in relationship to the epidemiologic database is assessed. These include, for example: (1) studies of respiratory tract deposition and clearance of particles; (2) experimental studies (animal and human) evaluating mechanisms of action of various particles (by size, chemical composition, etc.) in order to evaluate biological plausibility of effects reported by epidemiology studies; and (3) other experimental studies that demonstrate various toxic effects of PM constituents in humans or in animal models.

2.4.2.1 Respiratory Tract Dosimetry

The biological endpoint or health effect of an aerosol exposure is likely more directly related to the quantitative pattern of deposition within the respiratory tract than just to the external exposure concentration. The regional deposition pattern determines not only the initial respiratory tract dose but also the specific pathways and rates by which the inhaled material is cleared and redistributed. Thus, in order to evaluate different toxic responses to inhaled particles across species and to accurately extrapolate such laboratory animal data to humans, or to evaluate differences that sex, age, or disease may have on human variability, the various physicochemical, anatomic, and physiologic factors described must be integrated to estimate a deposited dose or perhaps a retained dose (deposition – clearance = retention). Delineation of the dose to each respiratory tract region (extrathoracic, tracheobronchial, and pulmonary) is desired because each region has different dominant factors controlling deposition and clearance, and different defense mechanisms. A theoretical model to describe particle deposition and clearance would require detailed information on all the influential parameters mentioned above (e.g., respiratory rates, exact airflow patterns, complete measurements of the branching structure of the respiratory tract, pulmonary region mechanics) for men, women, children, and across the various species used in toxicity studies. An empirical model (i.e., equations fit to experimental data) may adequately describe regional deposition and require much less data to develop the model structure.

Within the dosimetry chapter, the anatomy of the respiratory tract and the physicochemical, anatomical, and physiological factors controlling particle deposition, clearance, and retention are reviewed. Other factors that modify deposition, including sex, age, disease state, and exposure to irritants also are discussed. The available human and laboratory data on deposition and clearance and their positive and negative attributes for use in quantitative model development are discussed. Available validated model structures to estimate deposition and clearance in humans and laboratory animals are described and evaluated. The application of these models to quantitative extrapolation of the human and animal toxicity data also are discussed. Consideration is given to uncertainties in input parameters and the variability of model predictions when evaluating the usefulness of models for quantitative dose extrapolation.

2.4.2.2 Epidemiology Studies

Epidemiologic analyses are expected to provide some of the most crucial information useful in deriving health criteria upon which to base Agency decisions regarding possible revision of the current PM standards, and such studies are accorded extensive attention in this document.

One useful distinction is to separate short- and long-term PM exposure effects. The short-term effects include changes in respiratory function, symptom indicators, hospital admissions associated with exacerbation of respiratory or cardiovascular disease, and excesses of daily death rates in urban areas associated with concurrent 24-h PM measurements on the same or preceding few days. The short-term effects studies are typically longitudinal in nature and are specific to a community or metropolitan area with reasonably homogeneous PM exposures. The analyses of data in short-term studies use time-series analysis methods. The long-term or chronic exposure effects studies typically use annual PM concentrations and annual symptom or death rates and are more likely to involve comparisons across several communities rather than within a single community. Although both kinds of epidemiologic analysis are useful, it is important to assess the consistency of conclusions based on different kinds of studies. Coherence of effects at lower concentrations is a useful criterion for assessing diverse studies with different endpoints or effects, different populations, and different exposure metrics (Bates et al., 1990) and is considered as part of the evaluation of the available epidemiology literature.

Mortality Studies

Studies examining the relationship between ambient measures of PM and mortality were examined during the last review process (U.S. Environmental Protection Agency, 1982, 1986) and contributed to the key scientific bases underlying the current PM_{10} NAAQS. However, given the uncertainties in converting from British Smoke to PM_{10} measurements, particularly at lower concentrations, and the possible differences in particulate composition between London at the time the data were gathered and the contemporary United States, it was difficult to determine a precise level for a relationship between PM_{10} and mortality. Since that time, numerous contemporary U.S. mortality studies using either PM_{10} or TSP measurements have been published that examine short-term measurements. Also, long-term PM ambient measurements

and mortality have been examined in some recent studies. These and other newly emerging PM-mortality studies are summarized and critically evaluated.

Issues of greatest concern so far relate primarily to the use and interpretation of the short-term mortality studies. Almost all analyses of the relationship between PM and excess mortality require statistical adjustment for mortality excesses associated with other potential confounding factors, including other environmental stressors such as temperature and relative humidity or other pollutants (co-pollutants) associated with PM and with mortality. For example, weather-related effects may be directly related to excess mortality, but may also be indirectly related when weather affects PM emissions and atmospheric concentrations. Statistical and conceptual approaches to estimating the direct and indirect effects of covariates or confounders and specification of statistical adjustments for possible confounding factors are evaluated in interpreting the PM effects on mortality calculated from each study. Studies using different exposure metrics are considered and differences in particle size distribution or particle composition between cities are considered as the data allow.

Specification of "exposure-effect" relationship(s) between mortality and PM is also important. A number of studies have reported no evident threshold for effects, even at relatively low concentrations, but the ability to carry out meaningful threshold evaluations may be greatly limited by the statistical power of the available studies. Estimates of the relationship between PM and mortality may depend on differences in model specification. Even with similar model specifications (exposure-response relationship, adjustment for weather, copollutants, and other factors) there may be differences in the effects of PM at a given concentration, possibly related to differences in particle size/composition and/or climate or demographics among different cities. An important component of the health effects assessment in the criteria document is identification of susceptible subpopulations and other variables such as weather, climate, or other pollutants, potentially contributing to increased mortality risk.

Morbidity Studies

Decreased pulmonary function in predominantly healthy children was been reported in some earlier epidemiology studies. More recent studies add to this database. Earlier long-term exposure studies provided no evidence for an effect from PM exposure on level of pulmonary function, whereas some recent studies report reductions in pulmonary function associated with

chronic exposure to particulate pollution. An evaluation of the epidemiologic database relating short-term (24-h) and long-term (annual) ambient measurement of PM_{10} and other measures of PM to changes in pulmonary function test results in children and adults is presented. The strength and consistency of epidemiologic databases that relate short-term (24-h) and long-term (annual) PM_{10} and other ambient PM indicator measurements to changes in the rate and/or severity of respiratory symptoms and disease are also critically reviewed. Studies examining exacerbation of respiratory (i.e., COPD and asthma) and cardiovascular diseases that lead to increased medical care utilization (such as emergency room visits and hospital admissions) in relation to ambient PM exposure are also evaluated. As appropriate, other factors and copollutants are also examined in relation to findings on each of the above types of health endpoints.

2.4.2.3 Toxicology of Particulate Matter Constituents

In addition to assessing epidemiologic studies of PM differentiated mainly in terms of various size indicators (TSP, PM_{10}, etc.), the toxicology of various major subclasses of PM constituents is also evaluated. That evaluation focuses on acid aerosols, metals, ultrafine particles, diesel particles, silica, bioaerosols, and other types of particles that make up ambient air mixes of particles in the broad class designated in toto as "particulate matter". Animal inhalation toxicology and other types of studies are reviewed to ascertain information on several key health issues, e.g.: (1) the influence of particle size, number, and mass on health responses; (2) the differential influence of varying particle chemistry on the health effects observed; (3) the array of health effects that can be caused by specific PM constituents; (4) exposure-response relationships for various exposure durations (acute and chronic); (5) mechanisms of toxicity; and (6) pollutant interactions. Information from these studies relates to evaluation of the biological plausibility of the mortality and morbidity associations reported in epidemiological studies. The data on relationships among particle size, mass, number, and toxic effects may aid in determining the appropriateness of various exposure indicators of potential human effects.

Evaluation of the controlled human exposure (clinical) studies database concerning PM and health outcomes is presented as a subsection of the overall PM constituent toxicology chapter. This includes critical review of PM effects on pulmonary function in healthy and asthmatic individuals, pulmonary clearance mechanisms, airway reactivity, and immunologic

defense especially in relation to particle size but only to a limited extent in relation to chemical composition. There remains an almost complete absence of controlled experiment data on exposure of humans to particles other than acid aerosols.

Human clinical studies of PM constituents have been almost completely limited to measuring effects on symptoms, lung function, and airway reactivity, in addition to a few studies of effects on mucociliary clearance. Few have used bronchoalveolar lavage to study PM effects on airway inflammation and host defense; nor have many examined effects of acid aerosols or other particle exposures on airway inflammation in asthmatic subjects or on exacerbation of effects of antigen challenge in allergic or asthmatic subjects.

2.4.2.4 Sensitive Groups

Available data are also evaluated for insight concerning human population groups potentially having increased susceptibility to ambient PM exposure. Preexisting respiratory or cardiovascular disease, in conjunction with advanced age, appear to be important factors contributing to increased susceptibility to PM mortality. For morbidity health endpoints, children and asthmatic individuals may display increased sensitivity to PM exposure, and, as such, this topic is discussed.

2.4.3 Welfare Effects

2.4.3.1 Effects on Materials

All manmade materials exposed to the outdoor environment undergo degradation by heat, moisture, and some bacteria and fungi. For many years, air pollution has been suspected of accelerating the natural degradation processes. For example, acidic pollutants have been associated with accelerated degradation of paints such as water-based paint and alkyd coatings containing titanium dioxide, lead minium, or ferric oxide red. Other researchers have reported acidic pollution-related effects on automotive paint and steel coating. Particulate matter has also been reported to produce paint soiling. Also, acid aerosols and other particles containing acids also have been reported to affect building stones, cement, and concrete. Acidic or acid-forming aerosols change the physical characteristics of some stones, cement, and concrete by changing the chemical composition. Studies examining the effects on materials of PM pollution (primary

and secondary particles and aerosol precursor gases) are reviewed and summarized; where possible, changes in material damage are correlated with changes in PM concentrations.

2.4.3.2 Visibility Effects

There are several definitions for visibility; however, visibility is generally defined as the degree to which the atmosphere is transparent to visible light or a reduction in visual range and atmosphere discoloration. In 1977, Congress amended the Clean Air Act (CAA) to address problems with visibility impairment resulting from manmade air pollution, particularly in Class I Federal areas (national parks and wilderness areas). Airborne PM in the form of varying amounts of sulfates, ammonium and nitrate ions, elemental carbon and organic carbon compounds, water and smaller amounts of soil dust, lead compounds, and other trace species reduce visibility, thereby affecting transportation safety and creating a loss in aesthetic appeal. The fundamentals of visibility impairment, including the effects of PM concentration, aerosol composition, and size and pollutant emission trends on visibility are evaluated. Indicators of visibility and air quality are also discussed.

2.4.3.3 Climate Change

It has been suggested that fine particles released into the atmosphere may alter the climate through a reduction in the amount of solar radiation reaching the earth's surface, thus cooling the surface while heating the aerosol layer. The scattering and absorbing properties of aerosols and their vertical distribution are briefly reviewed and reference made to other assessments of their effects on radiative balance and how changes in radiative balance may affect weather and climate. Aerosols also affect weather and climate through their role as cloud condensation nuclei. The concentration, composition, size, and number of aerosols can influence the structure, stability, and albedo of clouds, possible changing the location and amount of rainfall and the rate of global and regional warming due to greenhouse gases. Airborne particles also play an important role in influencing the penetration of ultraviolet light (e.g., UV-B) to the surface of the Earth due to stratospheric ozone depletion, as is also briefly discussed.

2.4.3.4 Vegetation and Ecosystem Effects

Extensive information also exists which indicates that ambient PM (especially wet and dry deposition of acidic particles) can damage both terrestrial and aquatic vegetation and ecosystems. Such information is thoroughly evaluated elsewhere (Irving, 1991; U.S. National Acid Precipitation Assessment Program, 1991) but is not assessed in the present document.

2.5 DOCUMENT CONTENT AND ORGANIZATION

The present document critically reviews and assesses relevant scientific literature on PM through February, 1996. The material selected for review and comment in the text generally comes from the more recent literature published since 1982, with emphasis on studies conducted at or near PM pollutant concentrations found in ambient air. Older literature cited in the previous 1982 EPA PM AQCD and its Addenda (U.S. Environmental Protection Agency, 1982, 1986) is generally not discussed. However, as appropriate, some limited discussion is included of older studies judged to be significant because of their potential usefulness in deriving a NAAQS. An attempt has been made to discuss key literature in the text and present it in tables as well. Reports of lesser importance for the purposes of this document are typically only summarized in tables.

Generally, main emphasis is placed on consideration of published material that has undergone scientific peer review. However, in the interest of admitting new and important information, some material not yet fully published in the open literature but meeting other standards of scientific reporting may be included as reviewed by CASAC. As noted earlier, emphasis has been placed on studies in the range of current ambient levels. On this basis, studies in which the lowest concentration employed exceeded this level have been included if they contain unique data, such as documentation of a previously unreported effect or of mechanisms of effects, or if they were multiple-concentration studies designed to provide information on concentration-response relationships. In reviewing and summarizing the literature, an attempt is made to present alternative points of view where scientific controversy exists. As warranted, considerations bearing on the quality of studies are noted.

The present document consists of 13 chapters. The Executive Summary for the entire document is contained in Chapter 1, followed by this general introduction in Chapter 2.

Chapters 3 through 7 provide background information on physical and chemical properties of PM and related compounds; sources and emissions; atmospheric transport, transformation, and fate of PM; methods for the collection and measurement of PM; and ambient air concentrations and factors affecting exposure of the general population. Chapter 8 describes effects on visibility and climate, whereas Chapter 9 describes damage to materials attributable to PM. Chapters 10 through 13 evaluate information concerning the health effects of PM. More specifically, Chapter 10 discusses dosimetry of inhaled particles in the respiratory tract, and Chapter 11 summarizes information on the toxicology of specific types of PM constituents, including laboratory animal studies and controlled human exposure studies. Chapter 12 discusses epidemiological studies, and Chapter 13 integrates key information on exposure, dosimetry, and critical health risk issues derived from studies reviewed in the prior chapters.

Neither control techniques nor control strategies for the abatement of PM are discussed in this document, although some topics covered may be incidentally relevant to abatement strategies. Technologies for controlling PM emissions are discussed in other documents issued by EPA's Office of Air Quality Policy and Standards (OAQPS). Likewise, issues germane to the scientific basis for control strategies, but not pertinent to the development of criteria, are addressed in numerous other documents issued by OAQPS.

In addition, certain issues of direct relevance to standard setting are not explicitly addressed in this document, but are instead analyzed in documentation prepared by OAQPS as part of its regulatory analyses materials. Such analyses include (1) discussion of what constitutes an "adverse effect" and delineation of particular adverse effects that the primary and secondary NAAQS are intended to protect against, (2) exposure analyses and assessment of consequent risk, and (3) discussion of factors to be considered in determining an adequate margin of safety. Key points and conclusions from such analyses are summarized in a Staff Paper prepared by OAQPS and reviewed by CASAC. Although scientific data contribute significantly to decisions regarding the above issues, their resolution cannot be achieved solely on the basis of experimentally acquired information. Final decisions on items (1) and (3) are made by the Administrator, as mandated by the Clean Air Act.

A fourth issue directly pertinent to standard setting is identification of populations at risk, which is basically a selection by EPA of the subpopulation(s) to be protected by the promulgation of a given standard. This issue is addressed only partially in this document. For

example, information is presented on factors, such as preexisting disease, that may biologically predispose individuals and subpopulations to adverse effects from exposures to PM. The identification of a population at risk, however, requires information above and beyond data on biological predisposition, such as information on levels of exposure, activity patterns, and personal habits. Such information is included in the Staff Paper developed by OAQPS and reviewed by CASAC as a separate item from this document.

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